

IN THE SPECIFICATION

Please replace the title with the following new title:

Automated In-Process Ratio Mass Spectrometry for Trace Components of a Process Solution

Please replace the paragraph beginning on page 1, line 19 with the following replacement paragraph:

The present application is related to ~~an issued US Patent, No. 5,414, 259, issued to Howard M. Kingston on May 9, 1995, incorporated herein in its entirety by reference, to a co-pending patent application S/N 09/015,469, filed January 29, 1998, also incorporated in its entirety by reference, and to a provisional patent application filed January 29, 2001, bearing S/N 60/264748, now U. S. Serial No. 10/004,627 filed December 4, 2001, which is also incorporated herein in its entirety by reference..~~

Please replace the paragraph beginning on page 2, line 13 with the following replacement paragraph:

~~In a preferred embodiment the system of the invention uses a modified form of Isotope Dilution Mass Spectrometry (IDMS), known to the inventors as Speciated Isotope Dilution Mass Spectrometry (SIDMS).~~ The exemplary method is an elemental and speciation threshold measurement method that is optimized for quality assurance at and near instrumental detection limits. The threshold measurement method is automated for unattended operation, and describes an In-process, Atmospheric Pressure Interface Ionizer, Mass Spectrometer (IP-API-MS). The IP-API-MS apparatus is designed for identification and quantification of elemental contaminants or compounds and species in fluids, and in the exemplary case, liquid solutions.

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Please replace the paragraph beginning on page 4, line 17 with the following replacement paragraph:

In one embodiment, the ~~THE~~ present inventors have chosen an enhancement of a known technique for achieving calibration-free mass spectrometry measurements. The known technique is Isotope Dilution Mass Spectrometry, hereinafter IDMS. In IDMS, one takes advantage of well-known, naturally-occurring isotope ratios in essentially all elements. For example, it is well-known that the average atomic mass of copper (Cu) in the natural state is 63.546, consisting of isotopes 62.9396 at 69.2% and 64.9276 at 30.8%. Very generally speaking, assuming an analysis for Cu content is required, one takes a sample of the solution to be analyzed, spikes the sample with an enriched standard solution having a substantially different isotope ratio than the naturally-occurring ratio, introduces the spiked sample to a mass spectrometer, and records the measured mass ratio between the isotopes. The measurement is going to differ from the naturally-occurring ratio and the standard spike ratio, and from the measured ratio, knowing the quantity of the original sample, and the quantity of the spike solution, one can calculate the single unknown, that being the concentration of Cu in the original sample.

Please replace the paragraph beginning on page 5, line 29 with the following replacement paragraph:

It will be apparent to the skilled artisan that ~~employing the patented techniques of the above-referenced patent to Kingston is an excellent first step in accomplishing full-time, real-time mass spectrometry analysis of fluid systems, but that~~ there are many other challenges in sample collection, sample handling, sample spiking, dilution, control, and many other areas to accomplish such a robust measurement and control system in real applications, such as in wet-

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bath analysis in semiconductor manufacturing, which has been selected as an exemplary application for describing the apparatus and methods of the present invention..

Please delete the paragraphs beginning on page 6, line 15 through page 18, line 3 and replace with the following new paragraphs:

In accordance with the invention, an apparatus for in-process ratio mass spectrometry is provided. The apparatus includes: a spike dilution apparatus configurable to dilute a spike having a first concentration to produce a processed spike having a diluted second concentration; a mixer configured to mix the processed spike and an extracted sample having at least one analyte to permit equilibration therebetween; an atmospheric pressure ionizer (API) configured to ionize the equilibrated extracted sample and processed spike to produce ions; a mass spectrometer configured to process the ions by ratio determination; and a control system adapted to automatically control the spike dilution apparatus, the mixer, the API, and the mass spectrometer such the sample is mixed with the processed spike, ionized, and processed by the mass spectrometer, the control system being further configured to use the ratio measured by the mass spectrometer to characterize the concentration of the at least one analyte in the extracted sample.

In accordance with another aspect of the invention, a method of automatically analyzing an analyte is provided. The method includes the acts of: diluting a spike having a first concentration to produce a processed spike having a second concentration; wherein the second concentration is selected based upon an estimate of a concentration of the analyte in a sample; mixing the processed spike and the sample to produce an equilibrated mixture of the processed spike and sample; ionizing the equilibrated mixture using an atmospheric-pressure-ionizer to produce ions; processing the ions in a mass spectrometer to produce a response ratio; and characterizing the concentration of the at least one analyte using the response ratio.

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Please delete the paragraph beginning on page 18, line 31.

Please delete the paragraph beginning on page 19, line 11.

Please replace the paragraph beginning on page 19, line 17 with the following replacement paragraph:

~~Generally, the~~ The apparatus in embodiments of the present invention may use ~~uses~~ the ~~SIDMS~~ IDMS method for In-process measurement, using an Atmospheric Pressure Interface Ionizer coupled to a Mass Spectrometer (IP-API-MS). The IP-API-MS apparatus is designed for identification and quantification of elemental contaminants or compounds and species in fluids without reliance upon a high temperature argon plasma for equilibrium or requiring a high-pressure liquid chromatography HPLC separation step prior to measurement.

Please replace the paragraph beginning on page 32, line 8 with the following replacement paragraph.

Assume that a 33 ppb spike is required. The first step is the same as for the 1 ppm spike as described above, that is, 1 ppm spike is drawn from reservoir 611 into syringe 619. During this operation valve 613 may be opened to a controlled-pressure nitrogen source for the same purposes as previously described. While the 1 ppm solution is drawn into syringe 619, valves 641, 639, and 667 are set to connect syringe 645 to reservoir 669, and a precise volume of dilution solution is drawn into syringe 645. Now valve 667 is closed, and valves 641, 639, 637, and 635 are reset to straight-through, connecting syringe 645 to mixer 631 through an input conduit. At the same time, after syringe 619 is filled with the 1 ppm solution, valves 621 and 623 reset to connect syringe 619 to mixer 631 through another input conduit.

The arrangement of the input conduits are such that substantial fluid direction change will occur as syringes 619 and 645 are operated in concert, thereby aiding mixing. Syringes 619 and 645 are now operated in concert with syringe 643 with valves 625 and 627 set straight-through, and the volume of 1 ppm solution from syringe 619 is mixed with the dilution solution in syringe 645 through mixer 631 and into syringe 643. At the end of this operation accomplished through valve manipulation and manipulation of the appropriate syringe plungers, syringe 643 has a precise spike at 33 ppb.

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